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ON COMPLEX MAGNETIC SUSCEPTIBILITY OF A PARAMAGNETIC  
AT HIGH FREQUENCIES

by

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Summary

The thermodynamic theory of paramagnetic relaxation given earlier<sup>(1), (2)</sup> is used to determine the dependence, <sup>upon</sup> on constant transverse field, of the real and imaginary <sup>parts</sup> components of the complex magnetic susceptibility <sup>in the direction of an alternating</sup> (with reference to a variable field) of an ideal paramagnetic. ~~It is assumed that the para-~~ <sup>with</sup> ~~magnetic has pure spin magnetism; and that the frequency of the~~ <sup>alternating</sup> ~~variable field is large in comparison with the reciprocal of the~~ <sup>assumed to be</sup> spin-spin relaxation time. The experimental data<sup>(3)</sup> are considered from the point of view of the <sup>theoretical</sup> results obtained.

1. Suppose an ideal paramagnetic with pure spin magnetism is in an <sup>external</sup> external magnetic field  $\vec{H} = \vec{H}_0 + \vec{h}$  <sup>constant</sup> ~~not~~ the ~~permanent~~ component  $H_0$  of which is directed along the z-axis, while the variable portion  $\vec{h} \equiv \vec{h}^{\text{alt}}$  is along the x-axis (~~in~~ the case of perpendicular fields). Thermodynamic theory gives for the x-component  $\xi$  of the <sup>alternating</sup> ~~variable~~ portion of magnetization:

$$\xi = \chi \eta, \quad \chi = \chi' - i\chi'', \quad (1)$$

where

$$\frac{\chi'}{\chi_0} = \frac{(1 + \gamma_s^2 \omega_0^2)^2 + (1 - \gamma_s^2 \omega_0^2) \gamma_s^2 \omega^2}{[1 + \gamma_s^2 (\omega_0^2 - \omega^2)]^2 + 4 \gamma_s^2 \omega^2} \quad (2)$$

 $\omega$ 

omega

$$\frac{\chi''}{\chi_0} = \frac{[1 + \gamma_s^2(\omega_0^2 + \omega^2)] \gamma_s \omega}{[1 + \gamma_s^2(\omega_0^2 - \omega^2)]^2 + 4 \gamma_s^2 \omega^2} ; \quad (3)$$

$\chi_0$  is the static susceptibility (an ideal paramagnetic is being considered; therefore  $\chi_0 = b/T_0$ , where  $T_0$  is the lattice temperature, considered a constant, and  $b$  is the Curie constant);  $\omega_0 \equiv \gamma H_0$ , where  $\gamma$  is the ratio of the magnetic to the mechanical moment of the paramagnetic particles; and  $\tau_s = \tau_s(T_0, H_0)$  is the spin-spin relaxation time.

Phenomenologic theory <sup>(2)</sup> does not give the form of dependence of  $\chi'$  upon  $T_0$  and  $H_0$ , so that the dependence of  $\chi'$  and  $\chi''$  on  $T_0$  and  $H_0$  is seen to be only partially explained. However, one can give a dependence of  $\tau_s$  upon  $H_0$ , <sup>for</sup> ~~under~~ which theoretical formula <sup>(3)</sup> satisfactorily describes the experimental curve  $\chi'' = \chi''(H_0)$  (this curve is taken <sup>for</sup> with given  $T_0$  and  $\omega$ ); it turns out that  $\tau_s$  slowly increases with ~~growth of~~ <sup>and</sup>  $H_0$  with an asymptotic <sup>ally</sup> approaches ~~to the value~~  $\tau_s(\infty)$  for very large fields. ~~As a result of this~~ kind of variation of  $\tau_s$  with field, ~~it appears~~ <sup>would naturally mean</sup> that for sufficiently large frequencies (in the sense of  $\tau_s(\infty)\omega \gg 1$ ), it is possible to achieve the above-mentioned satisfactory description of the experimental absorption curve by simply assuming that  $\tau_s$  does not depend on  $H_0$ . In what follows we shall consider the <sup>oscillator</sup> ~~generator~~ frequency to be great enough in the above sense and shall determine the <sup>behavior</sup> ~~character~~ of the curves  $\chi' = \chi'(H_0)$  and  $\chi'' = \chi''(H_0)$  on this assumption.

2. Let us denote the <sup>ratio</sup> quantity  $\chi/\chi_0$  by B, and  $\chi''/\chi_0$  by A. Since we are now <sup>treating</sup>  $\gamma_0$  as independent of  $H_0$ , B and A are functions of  $H_0$  only through  $\omega_0$ ; we need to examine the nature of the relations  $B = B(\omega_0)$  and  $A = A(\omega_0)$  given by (2) and (3) for given  $T_0$  and  $\omega$ . In agreement with what was stated above we will consider that  $1/\gamma_0 \omega \equiv \epsilon \ll 1$ .

The function  $B(\omega_0)$  has a minimum and maximum accurate to terms of the first order in  $\epsilon$  at the points

$$\omega_{\min} = \omega - \rho_s \equiv \omega_0^{\min}, \quad \omega_{\max} = \omega + \rho_s \equiv \omega_0^{\max}, \quad (4)$$

respectively, and becomes zero at points

$$\omega_0 = \rho_s, \quad \omega_0 = \omega \quad (5)$$

where  $\rho_s \equiv 1/\gamma_0$ , so that  $\rho_s/\omega \equiv \epsilon \ll 1$ ; thus  $B(\omega_0^{\min}) < 0$  and  $B(\omega_0^{\max}) > 0$ . For the main terms of expression  $B(\omega_0^{\max})$  and  $B(\omega_0^{\min})$  we obtain

$$B(\omega_0^{\max}) = |B(\omega_0^{\min})| = \omega/4\rho_s. \quad (6)$$

For the main term  $B(0)$ , (2) gives for  $H_0 = 0$ :

$$B(0) = (\rho_s/\omega)^2, \quad (7)$$

and for  $H_0 \rightarrow \infty$  from (2) we obtain

$$B(\infty) = 1, \quad (8)$$

where supposing  $H_0 \rightarrow \infty$ , we intend to determine the <sup>behavior</sup> course of B for <sup>large</sup> high values of  $H_0$ , which however remain less than those values <sup>which</sup> ~~saturation of magnetization is closely approached~~ <sup>becomes saturated</sup> (since in the general theory <sup>[2]</sup> magnetization is assumed to be not too large). On the basis of (4), (6) and (8), instead of

(7) we may simply assume

$$B(0) = 0 \quad (9)$$

The curve  $A = A(\omega_0)$  has, accurate to terms of the third order in  $\epsilon$ , a maximum at the point

$$\omega_0 = \omega \quad (10)$$

and, accurate to terms of the first order in  $\epsilon$ , falls respectively half-way to the right and left of this point at the points

$$\omega_0 = \omega_{\min} \text{ and } \omega_0 = \omega_{\max} \quad (11)$$

For  $H_0 = 0$  and  $H_0 \rightarrow \infty$  from (3) we obtain respectively

$$A(0) = 1, A(\infty) = 0 \quad (12)$$

(the first is accurate to terms of the first order in  $\epsilon$ ). The curves  $A(\omega_0)$  and  $B(\omega_0)$  intersect at the point (if only the first terms are considered):

$$\omega_0 = \omega + \rho_B = \omega_{\max} \quad (13)$$

Similarly it is not difficult to see that the main term  $A(\omega)$  is equal to  $\omega/2\rho_B$ , so that (see (6)) we have approximately  $A(\omega) = 2B(\omega_{\max})$  in agreement with (11) and (13); and close to  $\omega_0 = \omega$  the curve  $A(\omega_0)$  is symmetrical and the curve  $B(\omega_0)$  antisymmetrical about this point.

3. The general <sup>form</sup> nature of the curves  $A(\omega_0)$  and  $B(\omega_0)$  is plotted in Figure 1.

Let us assume that the complex quantity  $\eta = h e^{i\omega t}$  indicates, for example, that the result of the measurement of the variable component of the magnetic field is  $\tilde{\eta} = \text{Im}(h e^{i\omega t}) = h \sin \omega t$ ; then the measurement  $\tilde{\xi}$  of the variable portion of magnetization

(1) gives

$$\bar{\xi} = \text{Im}(\chi\dot{\eta}) = \chi'\dot{\eta} - (\chi''/\omega)\dot{\eta}. \quad (14)$$

From the ~~character~~<sup>behavior</sup> of curves  $B(\omega_0)$  and  $A(\omega_0)$  it follows that for sufficiently high values for the constant field  $H_0$  (to the right of the point  $\omega_0 = \omega_0^{\text{max}}$ ), where  $\chi'' \ll \chi'$ , with <sup>respect</sup> reference to the variable field component there exists paramagnetism with magnetic susceptibility  $\chi > \chi_0$ :

$$\bar{\xi} \approx \chi \dot{\eta}, \quad (15)$$

while in the region of rather small values of the constant field (to the left of the point  $\omega_0 = \omega_0^{\text{max}}$ ), there is no simple relation between the variable part of the magnetic field and the variable part of the magnetization. In particular, when  $\chi'' \gg \chi'$  (this would be for very small values of  $H_0$ , and also for values of  $H_0$  close to  $\omega/\gamma$ ), we have the relation:

$$\bar{\xi} \approx -(\chi''/\omega)\dot{\eta}. \quad (16)$$

Zavoisky<sup>[3]</sup> was the first to propose a method for measuring the real component of the magnetic susceptibility of a paramagnetic at high frequencies. This method is based <sup>upon</sup> the anisotropy of the magnetic properties of a paramagnetic with <sup>respect</sup> reference to a variable field which occurs in the presence of a constant field, and upon the assumption of the independence of the parallel and perpendicular effects; i.e., <sup>the assumption</sup> of the independence of the complex magnetic susceptibilities with <sup>respect</sup> reference to an alternating (variable) field in directions parallel and perpendicular to a constant field (the correctness of this supposition can easily be seen to follow from the thermodynamic theory<sup>[1],[2]</sup> of paramagnetic relaxation). With his method Zavoisky obtained <sup>experimentally</sup>

the curve  $\chi'(H_0)$  for anhydrous manganese sulphate ~~the~~ wave length  $\lambda = 16$  cm. (i.e. ~~for~~  $\omega = 1.15 \cdot 10^{10}$  sec.<sup>-1</sup>). He also obtained experimentally ~~the~~ curve  $\chi''(H_0)$  for the same substance ~~the~~ same frequency. The ~~character~~ <sup>form</sup> of these curves is shown in Fig. 2 reproducing the general features of the corresponding diagram in Zavoisky's paper [3];  $\chi'$  and  $\chi''$  were measured in certain relative arbitrary units.

In ~~the~~ <sup>Zavoisky's</sup> work ~~the~~ [3] the following conclusions were drawn from the results of the measurements. First, the values for  $\chi'$  when  $H_0 \approx 0$  and at the point  $H_0 \approx \omega/\gamma$ , ~~for~~ which  $\chi''$  has its maximum, are identical; second, the minimum and maximum <sup>of</sup>  $\chi'$  are located at about those points where  $\chi''$  falls to half its value at maximum; third, within the limits of accuracy of the measurements, the differences in absolute magnitude between the values  $\chi'$  respectively at minimum and maximum and the value  $\chi'$  at the point  $H_0 = \omega/\gamma$  are equal; fourth, to the left of the point  $H_0 = \omega/\gamma$  there is diamagnetism with respect to the variable part of the field <sup>av</sup> to the right there is paramagnetism.

If in the case under consideration one may assume  $\tau_s \omega \gg 1$  (which appears likely, if  $\tau_s \approx 10^{-9}$  sec. [1], [2]), then Zavoisky's experimental results may be compared with the theoretical results obtained by us (although detailed comparison is considerably hampered by the circumstance that Zavoisky's measurements are only relative). Of the above listed conclusions drawn from the work discussed, the first agrees with our equations (5) and (9), the second with (4) and (11), the third with (6); as to the fourth conclusion, it is not quite correct.

Although  $\chi'$  does go through zero at the point  $H_0 = \omega/\gamma$ , this does not mean transition at this point from diamagnetism to paramagnetism with respect to the variable field; paramagnetism does actually begin near point  $H_0 = \omega/\gamma$  (to the right of point  $H_0 = \omega_0^{\max} / \gamma = \omega/\gamma + \rho_g/\gamma$ ), but generally speaking there is no diamagnetism in the sense of proportionality in magnitude but reversal in direction between the variable parts of the magnetic field and magnetization (see remarks relative to (14)-(16)). It would be very important for a more detailed comparison between the theoretical results and the experiment, to have available experimental data on the absolute values of  $\chi'$  and  $\chi''$ .

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#### References:

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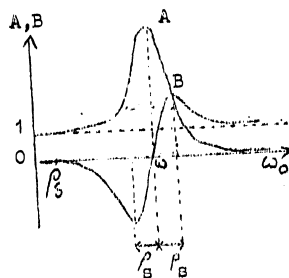


Figure 1.

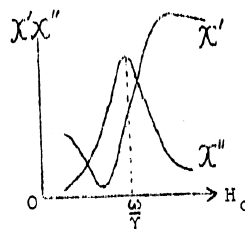


Figure 2.